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The Kerr nonlinearity of the beta-barium borate crystal

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A popular crystal for ultrafast cascading experiments is beta-barium-borate (β -BaB₂O₄, BBO). It has a decent quadratic nonlinear coefficient, and because the crystal is anisotropic it can be birefringence phase-matched for type I ($oo \rightarrow e$) second-harmonic generation (SHG). For femtosecond experiments BBO is popular because of low dispersion and a high damage threshold. The main attractive property of ultrafast cascading is that the induced cascading nonlinearity $n_{2,casc}^I$ can be negative, i.e. generate a self-defocusing Kerr-like nonlinearity. However, the material Kerr nonlinearity $n_{2,Kerr}^I$ is self-focusing and competes with the cascading nonlinearity. Therefore, precise knowledge of its strength is crucial. We perform an experiment measuring the main c_{11} tensor component, and together with literature experimental data [1], we propose a c_{11} value composed of 14 different data points.

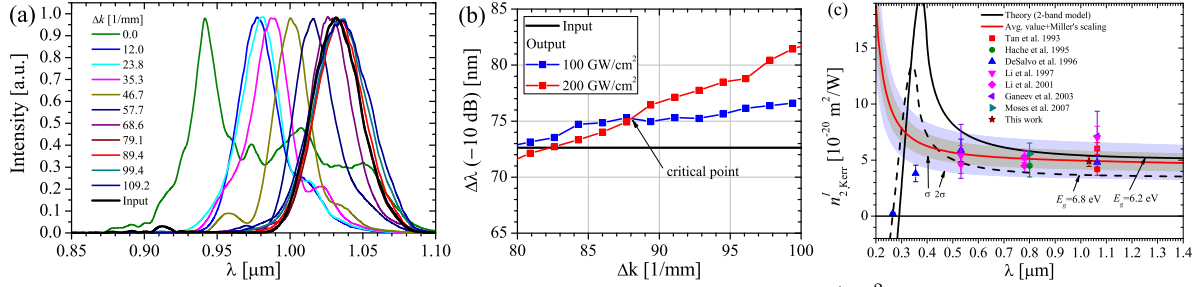


Figure 1. (a) Experimental spectra recorded with 50 fs@1030 nm and 200 GW/cm² transform-limited pulses, (b) the spectral bandwidth@-10 dB vs. Δk . (c) Summary of the experimental data from the literature corresponding to the c_{11} nonlinear susceptibility coefficient ($n_{2,Kerr}^I = 3c_{11}/4n_1^2\epsilon_0 c$). The data are corrected by us for cascading contributions. The shaded areas “ σ ” and “ 2σ ” represent one and two standard deviations.

BBO is a negative uniaxial crystal in the point group $3m$. When the pump is o -polarized, the nonlinear susceptibility component that accounts for the Kerr self-phase modulation (SPM) is $c_{11} = \chi_{XXX}^{(3)} = \chi_{YYY}^{(3)}$. We pumped a 25 mm BBO cut for $oo \rightarrow e$ SHG with 50 fs 1030 nm pulses from a commercial OPA. By tuning the phase mismatch away from zero cascading sets in as a Kerr-like SPM nonlinearity with the nonlinear index $n_{2,casc}^I = -2\omega_1 d_{eff}^2 / c^2 \epsilon_0 n_1^2 n_2 \Delta k$, where d_{eff} is the effective quadratic nonlinearity, and n_1 and n_2 the FW and SH linear indices. Fig. 1(a) shows that for low Δk the self-defocusing cascading dominates leading to strongly modulated and broadened spectra. At some critical point the cascading exactly cancels the material Kerr nonlinearity $n_{2,casc}^I + n_{2,Kerr}^I = 0$. Such a zero SPM nonlinearity should leave the spectra invariant with intensity. We found this point by observing the -10 dB bandwidth crossing of two different intensities, see Fig. 1(b). Then, by using the well-known quadratic nonlinearities for BBO we can calculate $n_{2,Kerr}^I = -n_{2,casc}^I = 4.9 \pm 0.4 \cdot 10^{-20}$ m²/W, which corresponds to $c_{11} = 4.7 \pm 0.4 \cdot 10^{-22}$ m²/V². In the literature other experiments have measured the Kerr nonlinearity in BBO [1]. We have done a careful analysis of these to (a) clarify which tensor components were excited, (b) ensure consistent definitions of the Kerr nonlinearity and (c) correct for the deterministic contribution from cascaded SHG [2]. The summary is shown for the most important c_{11} component in Fig. 1(c). The values agree surprisingly well with the two-band model, originally derived for wide-gap semiconductors. We confirmed that Miller’s delta, calculated as $\Delta_{11} = c_{11}/(n_1^2(\lambda_p) - 1)^4$, was nearly constant over all data at various pump wavelengths λ_p (except the UV measurements below 400 nm that were not used). Therefore we could calculate a weighted mean over 14 data points as $\Delta_{11} = 52.3 \pm 7.7 \cdot 10^{-24}$ m²/V², which corresponds to $c_{11} = 4.78 \cdot 10^{-22}$ m²/V² and $n_{2,Kerr}^I = 4.93 \cdot 10^{-20}$ m²/W at 1030 nm. Our experiment agrees very well with this average. We finally analyze literature data [3] to obtain the 3 other tensor components c_{10} , c_{16} and c_{33} as well.

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